SUSTAINABLE PROCESS ENGINEERING –SIR Group

EINDHOVEN UNIVERSITY OF TECHNOLOGY

Two-dimensional model in a packed bed membrane reactor for ammonia production considering diffusion limitation phenomena: optimization, design and scale-up

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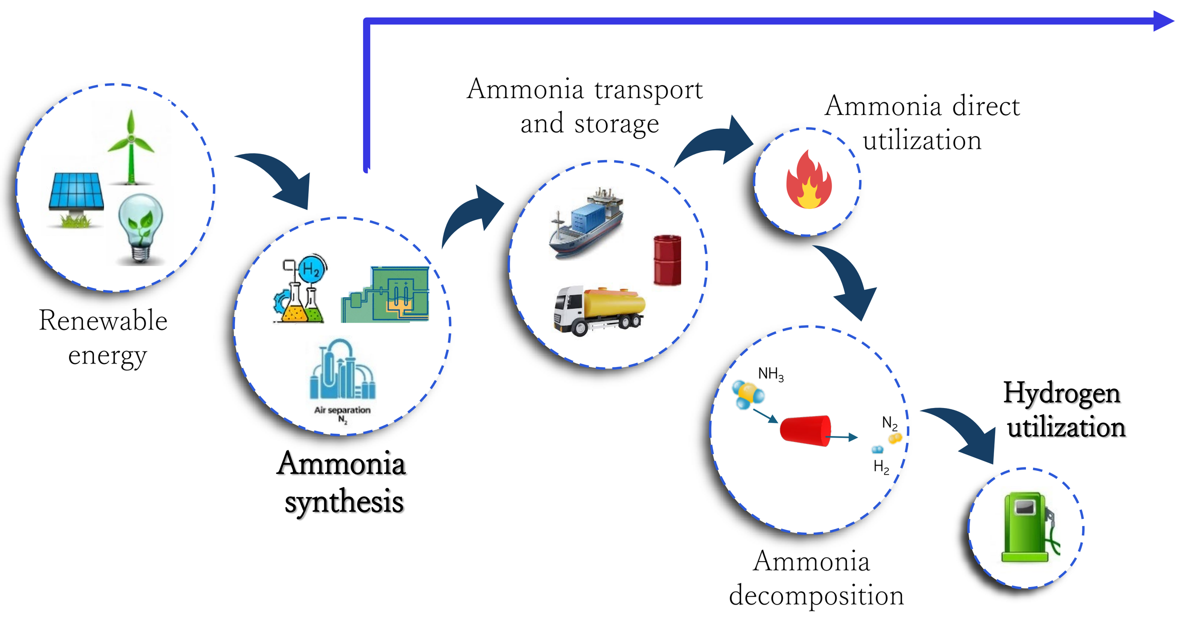
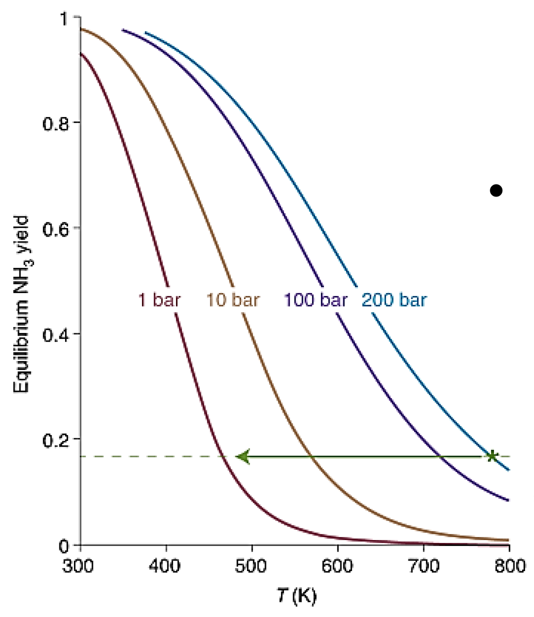
Introduction: NH_3 as new pathway for long-term energy storage



$N_2 + 3H_2 \leftrightarrow 2NH_3 \qquad (\Delta H_{298K}) = -90\ kJ/mol$



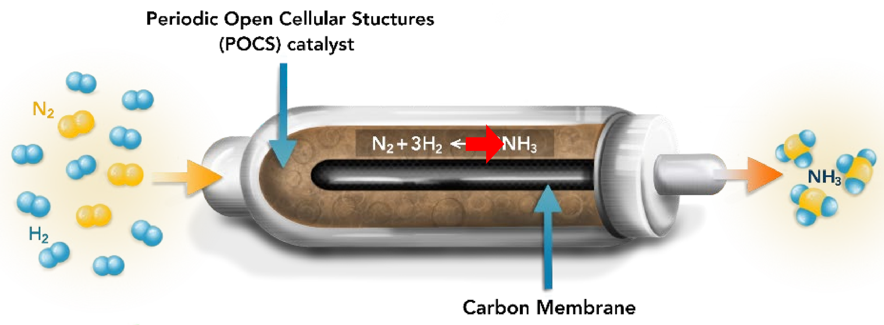
Challenges:

- **high inlet temperature** to achieve high reaction rate
- **high pressure** to achieve a high equilibrium conversion

Novel solution to overcome these limitations

Packed bed membrane reactor

for in-situ removal of NH_3, shifting the equilibrium conversion



- ✓ Effective heat management
- ✓ Energy efficiency
- ✓ Process intensification

Mathematical model: study of the packed bed membrane reactor (PBMR) phenomenology

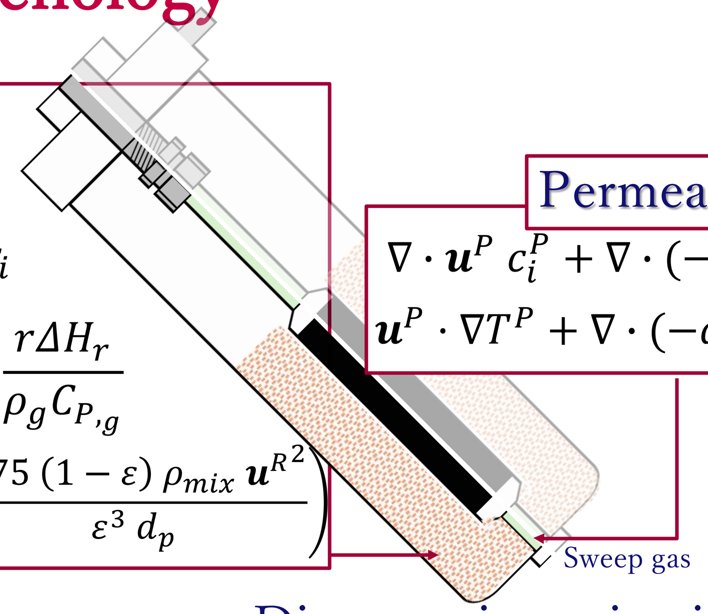
Retentate

- Steady-state total continuity equation: mass balance — $\nabla \cdot (\rho \boldsymbol{u}) = 0$
- Steady-state partial continuity equation: mass balance — $\nabla \cdot \boldsymbol{u}^R c_i^R + \nabla \cdot (-D_i \nabla c_i^R) = r_i$
- Steady-state partial continuity equation: energy balance — $\boldsymbol{u}^R \cdot \nabla T^R + \nabla \cdot (-\alpha^R \nabla T^R) = \frac{r \Delta H_r}{\rho_g C_{P,g}}$
- Steady-state partial continuity equation: momentum balance (Ergun equation) — $\frac{\partial P^R}{\partial z} = -\left(\frac{150\, \mu_{mix}\, \boldsymbol{u}^R (1-\varepsilon)^2}{\varepsilon^3 d_p^2} + \frac{1.75\,(1-\varepsilon)\, \rho_{mix}\, \boldsymbol{u}^{R^2}}{\varepsilon^3 d_p}\right)$

Permeate

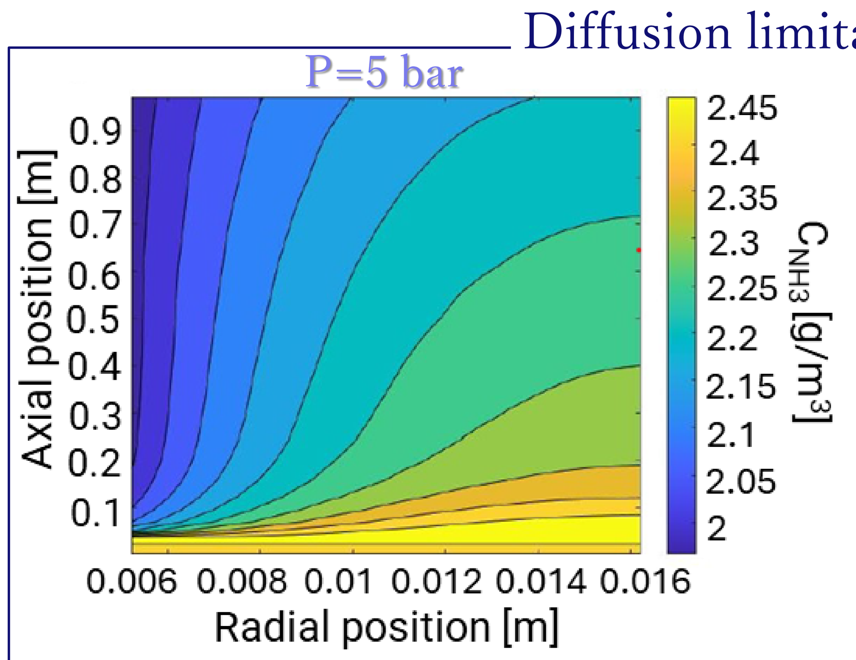
$$\nabla \cdot \boldsymbol{u}^P c_i^P + \nabla \cdot (-D_i \nabla c_i^P) = 0$$

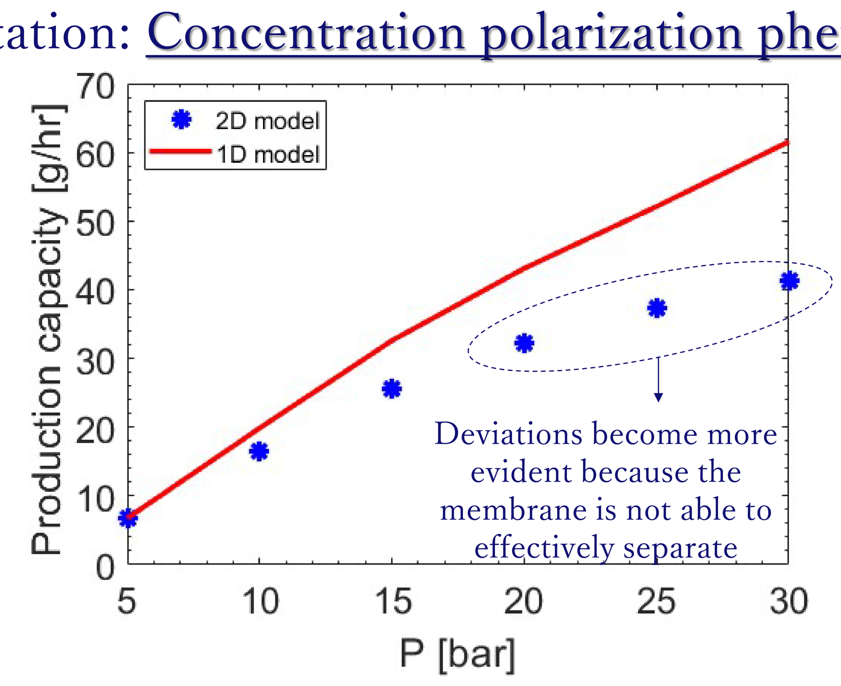
$$\boldsymbol{u}^P \cdot \nabla T^P + \nabla \cdot (-\alpha^P \nabla T^P) = 0$$

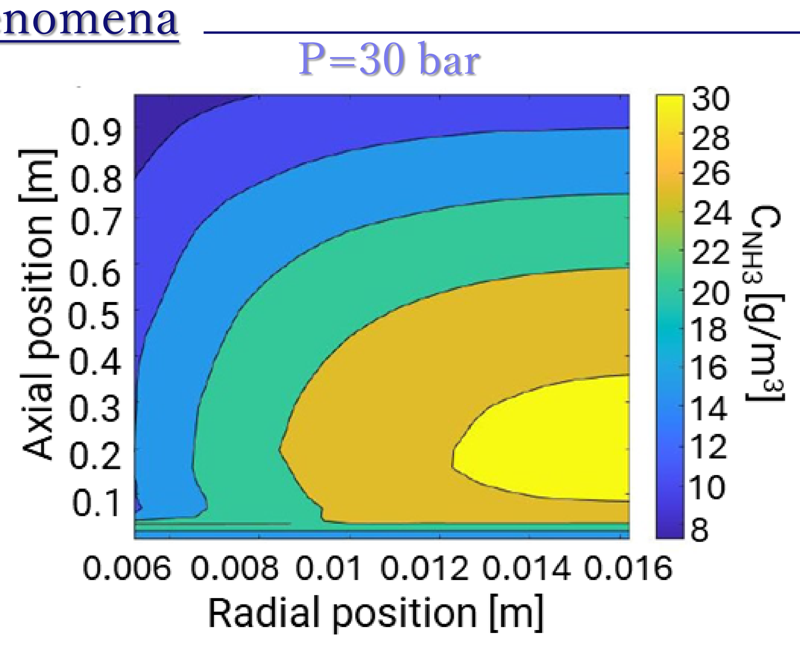


Simulation results: design and scaling-up solutions due to the diffusion phenomena

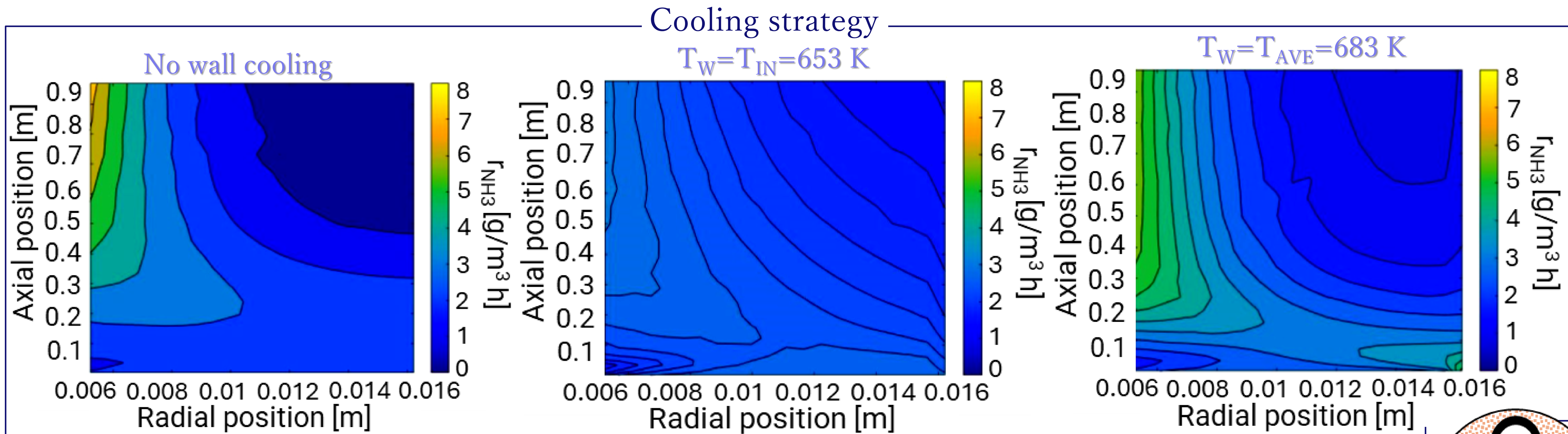
Diffusion limitation: Concentration polarization phenomena



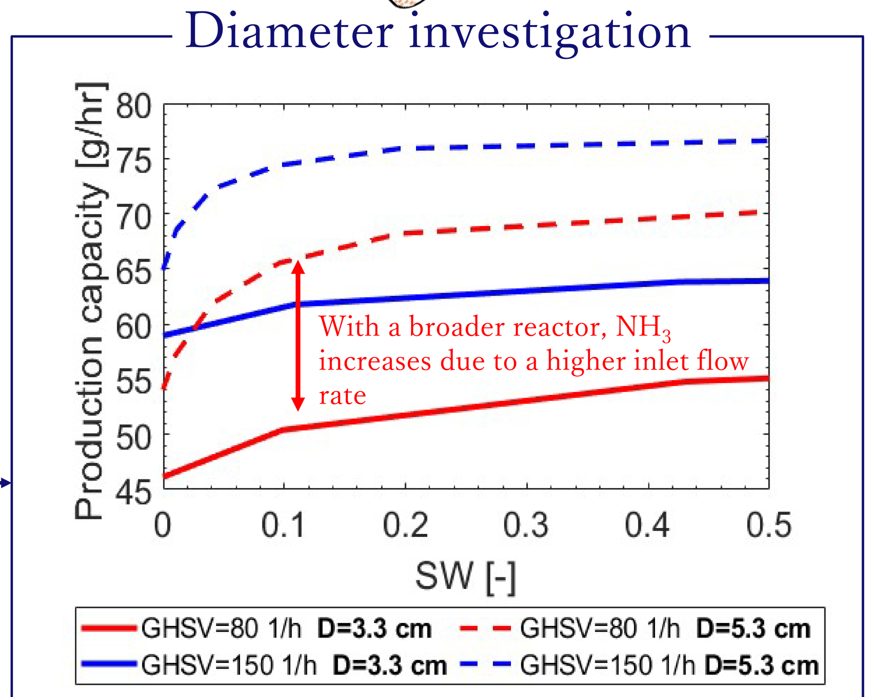


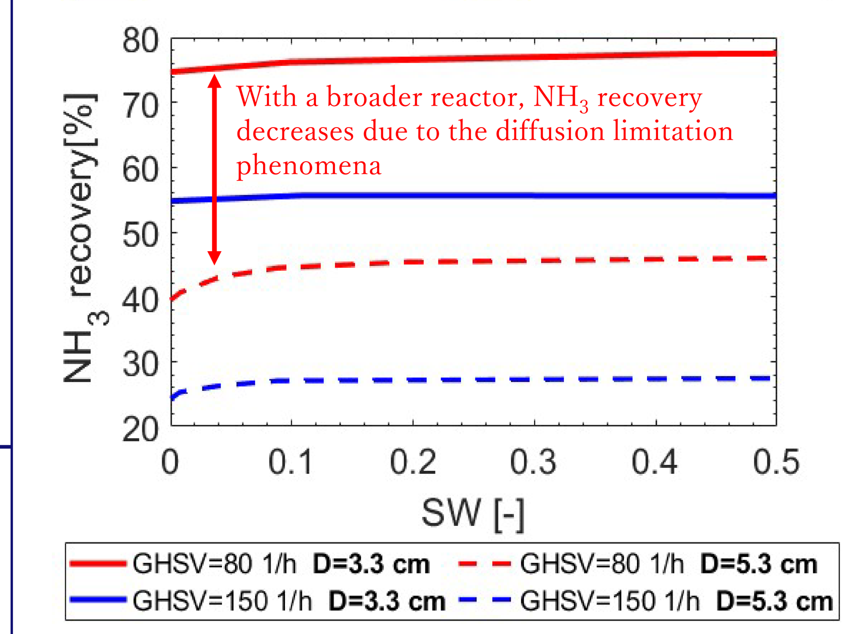


Cooling strategy

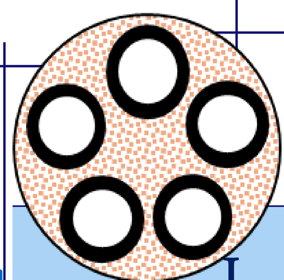


Diameter investigation





Multi-tubular membrane reactor to enhance membrane separation



Geom.	L_{mem} [m]	D_R [m]	Production capacity [g/hr]	Purity [%]	Catalyst weight [g]
1 mem	1	0.053	68.4	45.3	235
5 mem	1	0.053	62.5	34.3	175

Using 5 membranes of 0.2 m length each (total 1 m), the production capacity would be 8.6% lower than using one membrane of 1 m length, but it would still require 25% less catalyst

Conclusions

- **2D model** was **validated** with kinetic experimental test from literature and permeation lab experimental test
- **2D model outperform 1D** model even when $\frac{r}{L} \ll ReSc \ll \frac{L}{r}$. This is due to the diffusion limitation
- **Optimal** membrane reactor **conditions** were found for different geometries
- **Reactor length** study showed the advantage to have a multitube membrane reactor in terms of catalyst mass
- **Cooling strategy** study showed that too much low temperature slows down the kinetic

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